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PIXE and magnetic investigations of $\text{La}_x\text{RE}_{1-x}\text{F}_3$ (RE = Ce, Nd) single crystals

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ABSTRACT

The mixed $\text{La}_x\text{RE}_{1-x}\text{F}_3$ (RE = Ce, Nd) single crystals doped with Gd^{3+} (0.1 mol%) were grown by a modified Bridgmann-Stockbarger method. The PIXE (Proton Induced X ray Emission) method was used to the determination of a composition x of this material. The value of x for these samples does not differ more than 2.5 % and 7.5 % in $\text{La}_x\text{Ce}_{1-x}\text{F}_3$ and $\text{La}_x\text{Nd}_{1-x}\text{F}_3$ respectively, from the assumed one. In addition, concentrations of the doped rare earth atoms were controlled by XRF (X-Ray Fluorescence) method. The abundance of other than Gd^{3+} rare earth impurities was estimated to be below 0.058 at. %.

The magnetic susceptibility of the single crystals was measured in the temperature range 4 – 300 K, in a magnetic field $B = 0.2$ T applied in the crystallographic plane (001) perpendicular to the c -axis (along the a -axis), using the Faraday method. The magnetic susceptibility measurements confirm the lack of Ce^{3+} and Nd^{3+} clusters in the diluted crystals. The effective spins of hosts Ce^{3+} and Nd^{3+} are equal to 1/2 in the temperature range 4 – 300 K.

Keywords: rare-earth trifluorides, PIXE (Proton Induced X ray Emission), magnetic susceptibility, effective spin.

1. INTRODUCTION

The $\text{La}_x\text{RE}_{1-x}\text{F}_3$ (RE = Ce, Nd) single crystals doped with Gd^{3+} (0.1 mol%) were grown by a modified Bridgmann-Stockbarger method described elsewhere.^{1,2} Previously we studied defects and distortions of crystal cells in the same samples.^{2,3} Applying any technology to grow single crystals by cooling slowly the initially homogeneous liquid material (e.g. Bridgmann-Stockbarger method) one obtains a samples of different composition along the direction of crystallization. So it is important to know exactly a composition x of the investigated single crystals, which could be different from nominal one. Therefore we used the PIXE (Proton Induced X ray Emission) method in order to confirm the composition x . In addition the value x of the investigated samples was controlled using the XRF (X Ray Fluorescence) method. It is also important to know how the magnetic Ce^{3+} and Nd^{3+} ions are distributed on the lattice of the single crystals diluted with diamagnetic La^{3+} ions. For this purpose we used magnetic susceptibility method.

2. PIXE INVESTIGATION

The PIXE method was used to the determination of a composition x of the crystals. In this experiment X ray was induced by beam of 2.5 MeV protons from Van de Graaf accelerator in JINR, Dubna. The Si (Li) X ray detector with an energy resolution about 220 eV at 6.4 keV was used in that measurements. Typical PIXE spectra of the some investigated samples are presented in Figs. 1 and 2. Relationship between the concentration of RE (Rare Earth) was found by dividing of the corresponding squares for $K - X$ ray peaks, assuming equality of inducing efficiency and detection one. The composition x of the investigated samples does not differ more than 2.5 % and 7.5 % in $\text{La}_x\text{Ce}_{1-x}\text{F}_3$ and $\text{La}_x\text{Nd}_{1-x}\text{F}_3$ respectively, from the assumed one. In addition the composition x of the investigated samples was controlled using the XRF method. The abundance of other than Gd^{3+} rare earth impurities was estimated to be below 0.058 at. %. The results are summarised in Tables 1 and 2.

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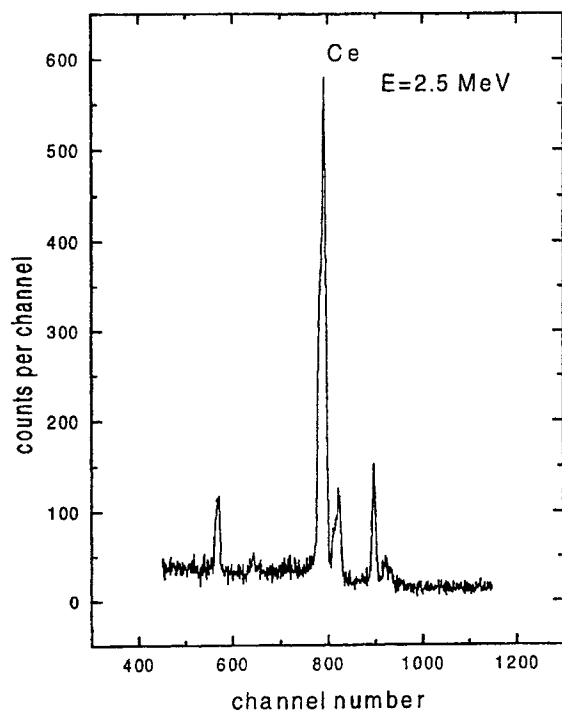


Fig.1. PIXE spectrum of the $\text{La}_{0.8}\text{Ce}_{0.2}\text{F}_3$ single crystal

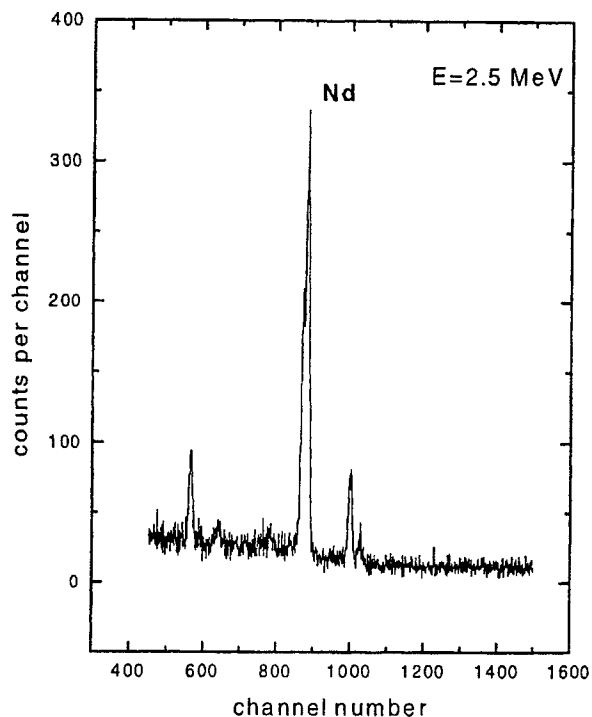


Fig.2. PIXE spectrum of the $\text{La}_{0.01}\text{Nd}_{0.99}\text{F}_3$ single crystal

Table 1. The composition of $\text{La}_x\text{Ce}_{1-x}\text{F}_3$ single crystals determined by PIXE method.

x	La (at.%)	Ce (at.%)
0.00	0.000	100.000
0.50	51.125	48.875
0.60	59.200	40.800
0.80	79.700	20.300
0.90	89.750	10.250
0.95	94.900	5.100
1.00	100.000	0.000

Table 2. The composition of $\text{La}_x\text{Nd}_{1-x}\text{F}_3$ single crystals obtained with the help of PIXE method.

x	La (at.%)	Nd (at.%)
0.00	0.000	100.000
0.01	1.075	98.925
0.02	2.100	97.900
0.05	5.225	94.775
0.10	10.400	89.600
0.90	89.600	10.400
0.95	94.825	5.175
0.98	97.935	2.065
0.99	98.970	1.030
1.00	100.000	0.000

3. MAGNETIC INVESTIGATIONS

The magnetic susceptibility of the $\text{La}_x\text{RE}_{1-x}\text{F}_3$ ($\text{RE} = \text{Ce}, \text{Nd}$) single crystals was measured in the temperature range from 4 K to 300 K using the Faraday method. In this experiment the magnetic field $B = 0.2$ T was applied in the crystallographic plane (001) perpendicular to the c -axis (along the a -axis). At these temperatures the single crystals remain paramagnetic for the all values of x in the range from 0 to 1.^{4,5} The crystal field effects and dipolar interactions between RE^{3+} ions influence significantly the magnetic susceptibility. The magnetic susceptibility of the investigated samples follows a Curie-Weiss law in the temperature range 100 – 300 K. The Curie -Weiss law parameters were given in our earlier works.^{4,5} Therefore the Curie constant C and the paramagnetic Curie temperature θ_p can be plotted as a function of x . The plots are presented in Figs. 3a and 3b. The Curie constant C and Curie-Weiss temperature θ_p scale linearly with x . It argues that the distribution of the magnetic Ce^{3+} and Nd^{3+} ions on the lattice is truly random – Ce^{3+} and Nd^{3+} host ions do not form the clusters. In addition magnetic study of the crystals revealed that the effective spins of hosts Ce^{3+} and Nd^{3+} are equal to 1/2 in the temperature range 4 – 300 K.^{2,4,5}

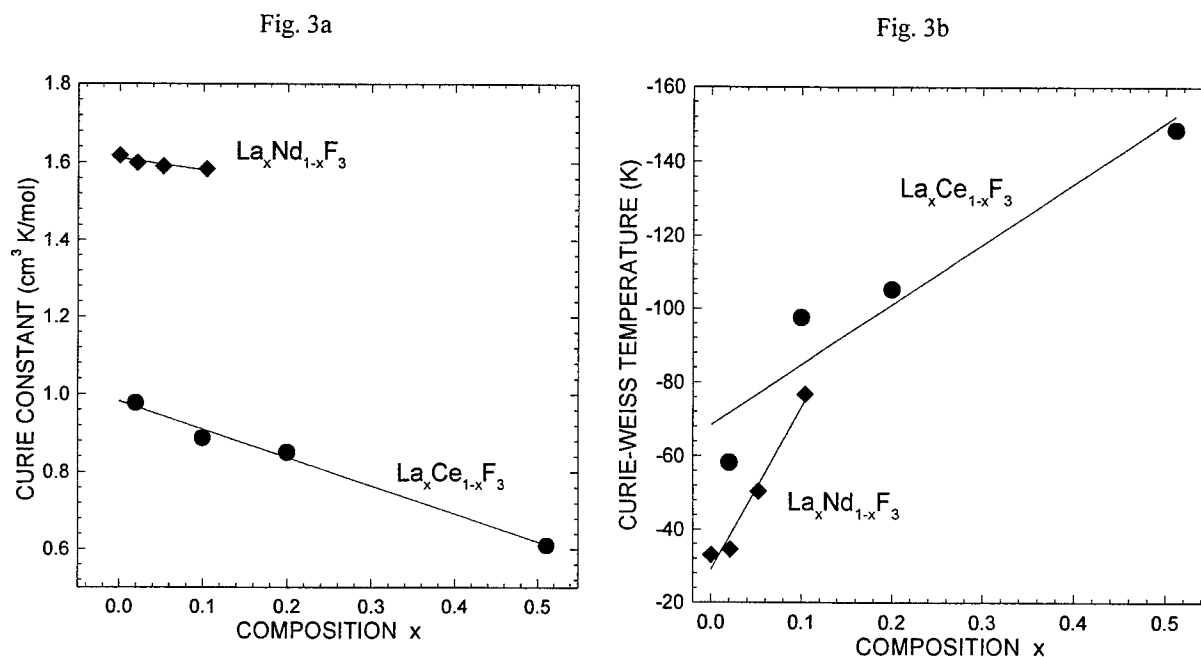


Fig. 3. The dependence of the Curie constant C and the Curie-Weiss temperature θ_p on composition x .

4. CONCLUSIONS

We have examined the composition x and the distribution of the magnetic Ce and Nd ions on the crystal lattice of the mixed $\text{La}_x\text{RE}_{1-x}\text{F}_3$ ($\text{RE} = \text{Ce}, \text{Nd}$) single crystals. The results confirm the good agreement with the earlier predictions of the composition and distribution of the magnetic Ce and Nd ions.

It can also be concluded that the effective spins of hosts Ce^{3+} and Nd^{3+} ions are equal to 1/2 in the investigated temperature range from 4 K to 300 K.

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